

Docket No.: 0071-0593PUS1  
(PATENT)

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of:  
Akira MITSUI et al.

Application No.: 10/500,701

Confirmation No.: 2439

Filed: July 2, 2004

Art Unit: 1711

For: LOW MOLECULAR WEIGHT  
POLYPHENYLENE ETHER

Examiner: D. Truong

**37 CFR § 1.132 DECLARATION**

***"Declaration I"***

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450  
Sir:

I, Akira Mitsui, a Japanese citizen, residing at 3-16-2, Fukuodai, Sodegaura-shi, Chiba 299-0261 Japan, do declare as follows:

I am the joint inventor of US application No. 10/500,701, entitled "LOW MOLECULAR WEIGHT POLYPHENYLENE ETHER";

I am familiar with the invention of the above-identified application and the prosecution history of US application No. 10/500,701;

I have read and understand the official action mailed on April 3, 2006 in the matter of US application No. No. 10/500,701 and the references cited therein (*i.e.*, Braat US '327 (US 6,211,327), WO '273 (WO 00/46273) and Dalton et al. US '164 (US 4,463,164));

In order to show and evidence that the invention of the above-identified application has

an advantage over the applied cited art, Examples and Comparative Examples (e.g., Comparative Example 5), which are reported in the present application were carried out by myself and/or in combination with my other co-inventors, and/or under our direction and control, and the results are described on pages 21-31 of the specification of the above-identified application.

I herein make the following true statements, in support of the fact that the present invention as claimed is distinguishable and patentable over the cited art references applied in the April 3, 2006 Office Action of the USPTO, and that the present invention has associated therewith unexpected and advantageous properties, when compared with the disclosures and teachings of such references.

*Statements*

In general, a process to produce PPE comprises following four steps:

*Step 1:* Polymerization of a monomer;

*Step 2:* Deactivation of a catalyst;

*Step 3:* Removal of a deactivated catalyst;

*Step 4:* Drawing of an obtained PPE.

In such a polymerization of PPE, a solvent is needed. In the present invention and the cited references being applied by the USPTO in the April 3, 2006 office action, three types of solvent appear, as is summarized in **Table I** immediately below.

Table 1

Solvent Type	Example of Solvent	Catalyst/ Monomer	Low molecular weight PPE		High molecular weight PPE
			$\eta_{sp}/c$ :	$<0.3$	$0.3<$
			Mw	$<25,000$	$25,000<$
A	Aromatic hydrocarbon (toluene, etc.)	Soluble	Soluble		Soluble
B	Aliphatic hydrocarbon (octane, etc.)	Soluble	Soluble		Insoluble
C	Alcohol (methanol, etc.)	Soluble	Insoluble		Insoluble

In the case where a Type A solvent is used alone as a polymerization solvent, the obtained PPE remains solved in the solvent at the end of above-mentioned Step 1. This method is called a “Solution Method”.

On the other hand, in case of using a Type B solvent, it depends on the desired molecular weight of PPE whether the obtained PPE is solved or not solved (i.e., precipitated). When the desired molecular weight of PPE is lower than 25,000 the obtained PPE is still solved in the solvent, and when the desired molecular weight of PPE is higher than 25,000 the obtained PPE cannot be solved, and thus is precipitated. This latter method is called a “Precipitation Method”.

Finally, in case of using a Type C solvent, the obtained PPE is precipitated, which is again called a “Precipitation Method”.

The low MW PPE of the present invention has a reduced viscosity of 0.04-0.18 dl/g, therefore, solvents of Type A and Type B are (good) solvents for the low MW PPE of the present invention, and a Type C solvent is a poor solvent (or “*anti-solvent*”) for the low MW PPE of the present invention.

Initial Points of Distinctions Over Braat US '327

The low molecular weight PPE of the present invention is distinguished from PPE obtained in the Braat US '327 reference. This fact is supported by comparative testing reported in the Specification.

In order to prove that the present invention has advantages over cited art being applied by the USPTO, and particularly the cited Braat US '327 reference, the USPTO's attention is directed to the instant specification at page 27, lines 8-24 "**Comparative Example 5**". In this respect, I submit that **Comparative Example 5** of the present application properly represents the PPE of Braat US '327, based on the following facts.

As described at page 27, lines 8-10 of the instant application:

*Comparative Example 5*

*According to the method described in the Example of U.S. Pat. No. 6,211,327,  
the reaction was carried out...*

In Comparative Example 5 as described at page 27 of the instant specification, PPE was obtained according to the method of Braat US '327 (*see page 27, lines 9-10 of the instant specification*). PPE obtained according to Comparative Example 5 (which is submitted to be equivalent to that obtained in the Example of Braat US '327) has a molecular weight distribution of 2.68 (see Table 1 at page 30 of the instant specification), while the molecular weight distribution of the present invention is 1.5-2.5. That is, the molecular weight distribution of the present invention is narrower (i.e., sharper) than that of Braat US '327.

Each of the reaction conditions of Comparative Example 5 in the present application and in the disclosed Example in Braat US '327 (see Braat US '327 Example beginning at column 8, line 50) shown in Table 2 immediately below.

**Table 2**

		<b>Comparative Example 5 of Present Application</b>	<b>Braat US '327 Example (Beginning column 8, line 50)</b>
<b>Step 1</b>	Monomer	2,6-dimethylphenol	2,6-xyleneol
	Solvent	Toluene	Toluene
	Catalyst	Cuprous bromide/ di-n-butyl amine (This has been missing at the translation. It can be seen in the priority document: which has been incorporated by reference )	Cuprous bromide/ di-n-butyl amine
	Temperature	40-45°C	40-45(48)°C
<b>Step 2</b>	Deactivator	Aqueous solution of nitrilotriacetic acid	Aqueous solution of nitrilotriacetic acid
	Heating	55°C, 70minutes	55°C, 70minutes
<b>Step 3</b>	separator	Liquid-liquid centrifuge	Liquid-liquid centrifuge
<b>Step 4</b>	Pre-condensation	Until the solid portion became 65%	Until the solid portion became 65%
	Condensation	using a devolatilization	using a devolatilization
		Solution was devolatilized	
<b>PPE</b>	ηsp/c	<b>0.112</b>	<b>0.113</b>
	Mw/Mn	2.68	(Not Explicitly Disclosed)
	Tg	162°C	(Not Explicitly Disclosed)
	Dielectric const.	<b>2.59</b>	(Not Explicitly Disclosed)

Thus, the PPE of Comparative Example 5 in the instant specification was obtained by using the same monomer, solvent and catalyst under the same conditions as were used in the Example of Braat US '373. Furthermore, treatment after polymerization was also carried out under the same conditions. Therefore, it is submitted that the obtained PPE in Comparative Example 5 of the instant application is substantially identical to the PPE obtained in the disclosed Example of Braat US '327 (which Example starts at column 8, line 50 thereof).

Accordingly, the fact that the PPE of Braat US'327 has a molecular weight distribution of 2.68 is well supported by Comparative Example 5 of the instant specification.

The USPTO has taken the position during prosecution of the present application that the molecular weight distribution of the present invention is inherent in Braat US '327, which is asserted to disclose the reactants and the steps of the instantly claimed process claims.

However, the methods disclosed in the present application obtain the product of the present invention, which product is distinguishable from the PPE product produced by the process of Braat US'327.

The low molecular polyphenylene ether of the present invention can be advantageously obtained, for example, by the method of pending claims 16-26. In the method of the present invention, recited features include the following (*e.g.*, see claim 16):

*"...precipitating a low molecular weight polyphenylene ether having a reduced viscosity (ηsp/c), as measured at 30 °C in a 0.5g/dl chloroform solution, of 0.04-0.18 dl/g by adding a poor solvent of said low molecular weight polyphenylene ether to said polyphenylene ether solution..."*

Namely, in method claim 16 (and product-by-process claim 27) precipitating a low molecular weight PPE by adding a poor solvent for said low molecular weight PPE to said

polyphenylene ether solution is one of the features of the method of the present invention used to prepare low molecular weight PPE of the invention.

In contrast, Braat US '327 *fails* to teach or disclose such a method or a low molecular weight PPE produced by such a method. For example, in Braat US '327 at column 2, lines 20-35, PPE is isolated through devolatilization of the reaction solvent (which is not a precipitation). Thus, this description in Braat US '327 means that the method of the cited Braat US '327 reference is a solution method, in which a precipitation of PPE would usually never substantially take place.

Further, it is known that PPE obtained by a solution method has a broader molecular weight distribution, since the polymerization of PPE take place continuously in a good solvent for PPE (see *Comparative Example 5 at page 27 of the instant application, which is substantially identical to Braat US '327 as explained above*).

This is quite different from the instant methods of the present invention where a poor solvent is used to precipitate the low molecular weight PPE.

More particularly, the disclosure at column 6, lines 48 and 37-38 of Braat US '327 corresponds to **Steps 2 and 3** of a typical process to produce PPE (*i.e.*, **Step 2: Deactivation of a catalyst, and Step 3: Removal of a deactivated catalyst**) as explained above.

That is, in Braat US '327 the purpose of adding an *anti-solvent* in combination with water is not to precipitate PPE, but is instead to make the catalyst easily removable. In other words, a water-phase including catalyst and an oil-phase including PPE can be separated easily by adding an *anti-solvent*. This is a typical approach employed in a solution method. Thus, in Braat US '327, precipitation does not take place even if an *anti-solvent* in combination with water is added in its process.

The temperature at the time of precipitating PPE also affects the polymerization, as is shown in the Examples in the instant specification (i.e., see Examples 9 and 10, and Comparative example 6). In order to obtain the low MW PPE in a high yield through the inventive method recited in claim 16 (and claim 27), the temperature at the time of precipitating the PPE is significant.

There is no disclosure in the cited art of record, which is concerned with and/or is related to the temperature at the time of precipitating PPE. The cited Braat US '327 reference only discloses the temperature at the time of polymerization and not at the time of precipitation. In the polymerization progress of PPE, polymerization usually is carried out at the temperature of around 40 °C, and the process after polymerization, that is Steps 2 - 4, are also carried out under a temperature that is the same as with polymerization. Thus, Braat US '327 fails to give any consideration to the temperature at the precipitation.

#### Unexpected Results Associated with the Present Invention

Except as indicated below for values reported for Dalton US '164 in Figure 1, the test procedures used to gather the data set forth in Exhibit Figures 1-3 of this declaration are described in the instant specification, and the test data reported in Exhibit Figures 1-3 for Examples and Comparative Examples are from the same Examples and Comparative Examples reported in the instant specification.

Data relating to Dalton US '164 in Figure 1 that is reported in Figure 1 was obtained by calculating a specific  $\eta_{sp}/c$  value based on Mw values disclosed in Dalton US '164 (i.e., the disclosed Dalton US '164 Mw was converted to an  $\eta_{sp}/c$  value using a formula for conversion to Mw from  $\eta_{sp}/c$ , which is as follows:  $Mw = 112000 \times (\eta_{sp}/c)^{1.26}$ .



The relationship between the present invention, Braat US '327 and Dalton US '164 is shown in *Exhibit Figure 1* enclosed herewith. From Exhibit Figure 1, it is clearly realized that the present invention is distinguished from the cited references in characteristics such as Molecular Weight Distribution (MWD) and viscosity.

More specifically, in **Exhibit Figure 1** attached hereto, it is clearly shown that the PPE of the present invention has a very distinct viscosity ( $\eta_{sp}/c$ ) and Molecular Weight Distribution (MWD) as compared with compositions of Comparative Example 5 in the specification (which is equivalent to the Braat US '327 reference), Comparative Examples 1 and 2 of the instant application, and Examples 1, 5, 6, 7 and 8 of Dalton US '164).

Similarly, in **Exhibit Figure 2**, attached hereto, it is shown that Examples of the present invention (Examples 1, 3, 7, 8, 9a, 9b and 10a) , have a distinct high thermal resistance  $T_g$  ( $^{\circ}\text{C}$ ), at a change of viscosity ( $\eta_{sp}/c$ ), which is not possessed by either Comparative Example 5 (which is equivalent to the Braat US '327 reference), or Comparative Examples 1 and 2 of the instant application. More particularly Exhibit Figure 2 shows the present invention ( $\blacktriangle$  and  $\blacksquare$ ) has a higher thermal resistance ( $T_g$ ) than Braat US '327 ( $\square$ ), at the same viscosity.

**Exhibit Figure 3** attached hereto shows the dielectric constants. In Exhibit Figure 3, it is shown that the present invention ( $\blacktriangle$  and  $\blacksquare$ ) has results better than Comparative Example 5 of the present specification (which is equivalent to the Braat US '327 reference ( $\square$ )) at the same viscosity ( $\eta_{sp}/c$ ).

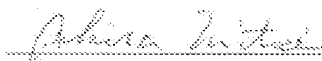
Accordingly, the comparative test results provided in Exhibit Figures 1-3 and the facts of distinction set forth above, evidence that the PPE of the present invention as recited in the

pending claims is both patentably distinct from and non-obvious over the disclosures of the cited art of record.

This conclusion is based on the fact that none of the cited references (*i.e.*, Braat US '327 (US 6,211,327), WO '273 (WO 00/46273) or Dalton et al. US '164 (US 4,463,164)) provide any teaching or disclosure that would allow one to arrive at the instant inventive PPE as presently claimed, which possesses unexpected and advantageous properties not possessed by products of the cited art applied by the USPTO in rejecting the pending claims in the April 3, 2006 office action of record.

The undersigned declarant declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

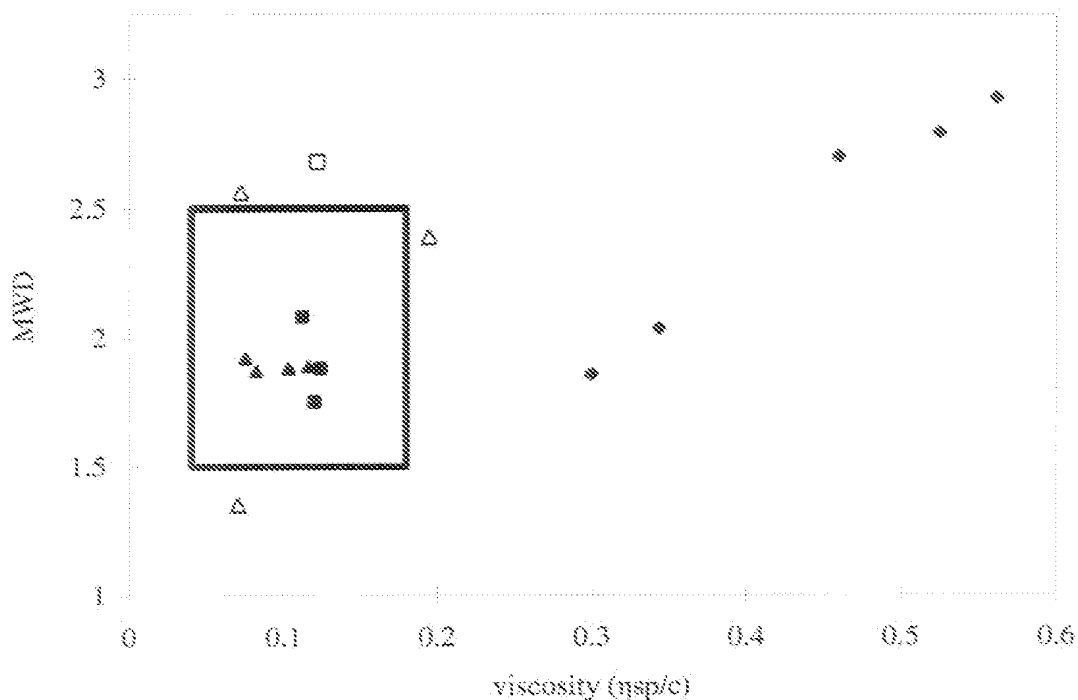
Signed this 21 day of September 2006.



Akira Mitsui

Attachments: Exhibit Figures 1-3

Exhibit Figure 1



MWD stands for Molecular Weight Distribution.

$\square$  The **Bolded Square/rectangle** represents the scope of viscosity and molecular weight distribution of the present invention.

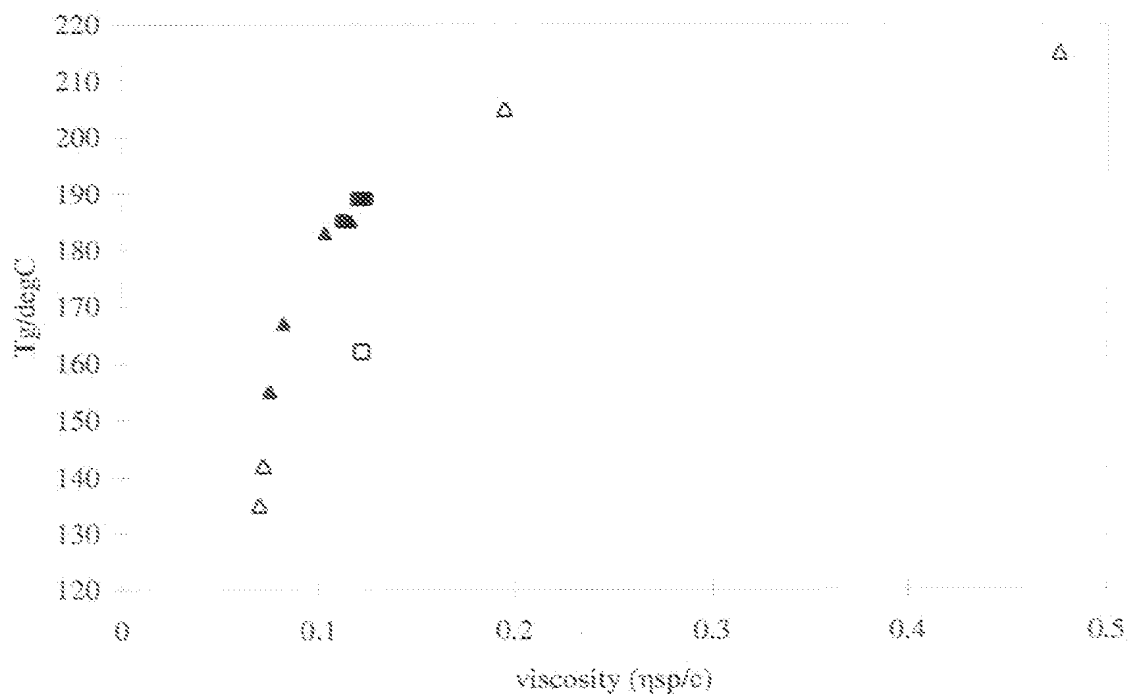
$\Delta$  and  $\blacksquare$  represent Examples of the present invention (Examples 1, 3, 7, 8, 9a, 9b and 10a).

$\square$  represents Comparative Example 5, equivalent to the Braat US '327 reference, of the instant application.

$\Delta$  represents Comparative Examples 1 and 2 of the instant application.

$\blacklozenge$  represents Dalton US '164 (Examples 1, 5, 6, 7 and 8 of Dalton US '164).

Exhibit Figure 2

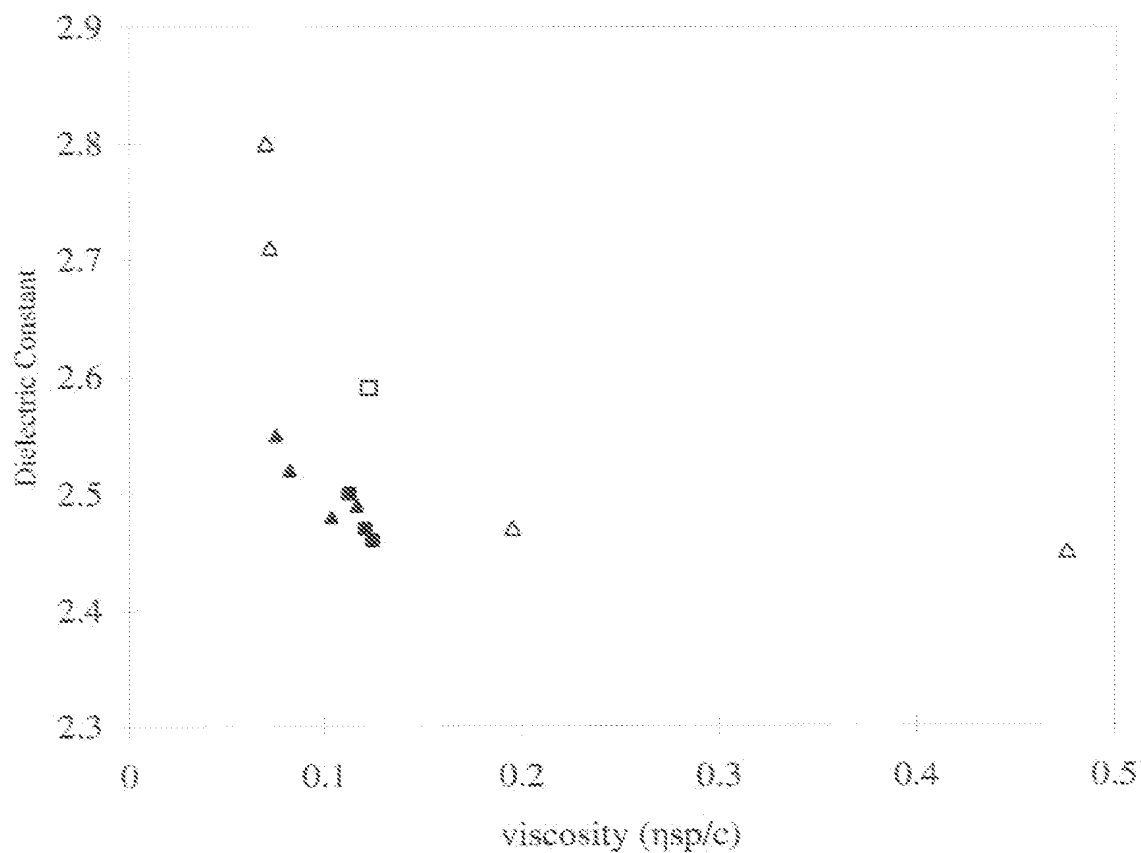


▲ and ■ represent Examples of the present invention (Examples 1, 3, 7, 8, 9a, 9b and 10a) .

□ represents Comparative Example 5 of the instant application (which is equivalent to the Braat US '327 reference).

△ represents Comparative Examples 1 and 2 of the instant application.

**Exhibit Figure 3**



▲ and ■ represent Examples of the present invention (Examples 1, 3, 7, 8, 9a, 9b and 10a).

□ represents Comparative Example 5 of the instant application (which is equivalent to the Braat US '327 reference).

△ represents Comparative Examples 1 and 2 of the instant application.